

REMARKS

This document is submitted in reply to the Office Action dated November 13, 2007 (Office Action).

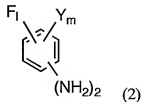
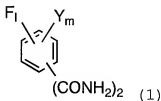
Applicants have amended claim 1, 5, 6, 11, 12 to more particularly point out their invention. Support for the amendment to claim 1 can be found in the Specification at page 8, lines 25 and 26. Support for the amendment to claims 5, 6, 11, and 12 can be found in original claim 1. More specifically, Applicants have incorporated all the limitations of claim 1 into claims 5, 6, 11 and 12. Applicants have also amended claim 3 to correct a typographical error. Upon entry of these amendments, claims 1-12 will be pending and under examination. No new matter has been added.

Applicants respectfully request that the Examiner reconsider this application, as amended, in view of the following remarks.

I

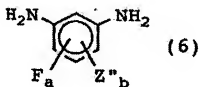
Claims 1-12 are rejected as obvious over Masayoshi et al., European Patent Application No. 1275679 (Masayoshi), in view of Hazen et al., U.S. Patent No. 5,011,997 (Hazen). Independent claim 1 will be discussed first.

Claim 1, as amended, covers a method for obtaining a fluorinated phenylenediamine of formula (2) shown below. The fluorinated diamine (2) is obtained via a Hoffman rearrangement by reacting a diamide, represented by formula (1) below, with NaOX, where X is either Br or Cl, in the presence of NaOH. In this method, the molar ratio of NaOX to diamide is in the range of 3.0 to 6.0.¹



¹ Claim 1 recites the additional limitation of "NaOH to diamide is in the range of 1.8 to 6.0." As will be discussed below, the patentability of claim 1 resides in part in "a NaOX/diamine ratio within the range of 3.0-6.0," not "a NaOH/diamine ratio within the range of 1.8-6.0."

As correctly pointed out by the Examiner, Masayoshi, the primary reference, discloses “a fluorinated phenylenediamine of ... formula (6) [shown below] where Z” denotes a chlorine, bromine or iodine ... a denotes fluorine atoms bonded to a benzene ring, representing an integer of 0-4 ... and b denotes the number of ‘Z”’ bonded to a benzene ring, representing 0-4 ... [such that] a and b ought to be invariably 4.” See the Office Action, page 2, line 16 through page 3, line 5.



As also correctly pointed out by the Examiner, “Masayoshi does not teach a method for the production of a fluorinated aromatic diamine from [a] corresponding diamide.” See the Office Action, page 3, last sentence. Thus, Masayoshi does not teach a method for obtaining a diamine from a corresponding diamide, wherein the molar ratio of NaOX to diamide is in the range of 3.0 to 6.0 as required by claim 1.

Turning to Hazen, the secondary reference, the Examiner states that “Hazen discloses a method for the production of a fluorinated aromatic diamine which comprises steps of reacting a diamide with NaOCl at a molar ratio of the NaOCl to the diamide in the range of 2.1-2.2 and NaOH.” See the Office Action, page 4, lines 1-5. In the Hazen method, the NaOX/diamide ratio is in the range 2.1-2.2, not of 3.0-6.0 as required by claim 1. Applicants submit that one of ordinary skill would not have been motivated to increase this ratio, in view of Hazen’s teaching that “[h]igher excesses of the hypohalite ... will increase the amount of azo compound formation ...” See column 4, lines 27-30. As will be discussed below, Hazen clearly teaches away from the higher NaOX/diamide ratio of amended claim 1 judging from the just-quoted teaching about increased formation of an azo by-product.

Applicants would like to point out that to increase diamine yield and purity, the Hazen method relies on a second step in which the azo by-product generated during the

Hoffman reaction is subjected to a hydrogenation reaction using nickel or nobel metal catalysts. See column 2, lines 8-11. This second step is key to the Hazen method. As pointed out in Hazen, “[t]he first step of the process known as the Hoffmann Reaction for the conversion of an amide to the primary amine ... does not produce a high purity product in high yield but rather a low purity product which is extremely difficult to purify.” See column 2, lines 3-8.² In other words, the azo by-product contributes to diamine impurity and low yield during the Hoffmann reaction. Thus, it has to be removed, e.g., via hydrogenation reaction. Accordingly, one of ordinary skill in the art, in view of the teachings in Hazen, would have been motivated to decrease, rather than increase, the NaOX/diamide ratio as increasing this ratio would result in higher amounts of azo by-product thereby rendering the purification more difficult.³ In short, Hazen teaches away from a NaOX/diamide ratio in the range of 3.0-6.0, as required by amended claim 1.

In sum, Masayoshi, in combination with Hazen, does not render the method of amended claim 1 obvious as neither reference teaches a NaOX/diamide ratio in the range of 3.0-6.0. Nor do they render obvious claims 2-4 and 7-10, which depend directly or indirectly from amended claim 1.

Applicants would like to bring to the Examiner’s attention that even if a prima facie case of obviousness had been established based on Masayoshi and Hazen (which Applicants do not concede), it can be successfully rebutted by a showing of unexpected results.

² The Examiner alleges that, “Hazen teaches [] the above Hoffmann rearrangement process results in a product of high purity (more than 95 %) and high yield [above 90 %], which is much higher compare[d] to ones of traditional methods.” See the Office Action, page 4, last sentence through page 5, line 1. As discussed above, it is the second step of purification, not the Hoffmann reaction, which results in increased diamine yield and purity.

³ In reference to the NaOH/diamide ratio, it is the Examiners position that “in [the] Hazen process a molar ratio of the NaOH to the diamide (NaOH/diamide ration) is higher than 6.0.” See the Office Action, page 4, lines 6-11. In other words, the Examiner’s stance is that Hazen does not teach or suggest a molar ratio of NaOH to diamine in the range of 1.8 to 6.0, as required by amended claim 1. Applicants agree with the Examiner. See footnote 1.

Examples 1-3 of the Specification (pages 23-26) and a new Example described in a declaration by Yasunori Okumura, copy attached hereto as "Exhibit A," show unexpected results. As shown in these four examples, a significant increase in diamine yield and purity was achieved by keeping the NaOX/diamide ratio within the range of 3.0-6.0. In these examples, the NaOX/diamide ratios were within the range of 3.0- 4.0 and resulted in yields within the range of 63.0 % to 80.7 %. These yields are significantly greater than the yields of 16.4% and 4.3% in Comparative Examples 1 and 3, respectively. The corresponding NaOX/diamide ratios were 2.0 and 2.5 in these two comparative examples. Of note, the NaOX/diamide ratios of 2.0 and 2.5 in Comparative Examples 1 and 3 are similar to those taught in Hazen i.e., 2.1-2.2. Furthermore, the diamine purity shown in Examples 1-3 was in excess of 99.8 %, vastly higher than that described for the diamine product of the Hazen method, and as a result, there was no need to conduct a hydrogenation reaction to remove the azo by-product as taught in Hazen.

Applicants now turn to the rejections of amended independent claims 5 and 11. Claims 5 and 11, as amended, each cover a method for obtaining a polyamic acid. The first step in each method is identical to that of amended claim 1, discussed above. Thus, for the same reasons and facts set forth above for claim 1, amended claims 5 and 11 are also not rendered obvious by the combination of Masayoshi and Hazen. Nor are claims 6 and 12 which depend from claims 5 and 11, respectively.

II

Claims 1-12 are rejected as obvious over Masayoshi, in view of Andrews et al., Aust. J. Chem., 1971, 412-422 (Andrews). Independent claim 1 will be discussed first.

As discussed above, Masayoshi does not teach or suggest a method for obtaining a diamine from a corresponding diamide let alone a method where the NaOX/diamide ratio is in the range of 3.0 to 6.0, as required by claim 1.

Turning to Andrews, the secondary reference, the Examiner correctly points out that, "Andrews discloses preparation of Isophthalic-based diamines from corresponding [diamides] with [the] Hoffmann rearrangement." See the Office Action, page 4, lines 12

and 13. According to the Andrews method, NaOBr is generated by in situ by reaction of bromine and NaOH. This reaction, well known in the art, is shown below:⁴



In order to determine the NaOX/diamine ratio, one must first determine the number of mmols of NaOBr generated, in accordance with the above formula. The calculation for the diamine product 1, 3-diamino-2-nitobenzene, disclosed in Andrew at page 416, lines 22, is as follows:

Diamide used in this example is **12.0 mmols**. Bromine (Br₂) used in this example is 24.0 mmol. NaOH used in this example is 75.0 mmols. 24.0 mmols of Br₂ reacts with twice the number of mmol of NaOH. Thus, the number of mmols of NaOH used in this reaction is 48.0 mmols. This reaction produces **24.0 mmol** of NaOBr. The amount of the other two products NaBr and H₂O formed are 24.0 mmol each. Upon completion of the reaction, 27 mmols of NaOH remains, i.e., 75.0 mmol (initial) minus 48.0 mmol (consumed).

Thus, the NaOBr/diamide ratio, disclosed by Andrews, is **2.0** (i.e., 24.0 mmols NaOBr/12 mmols diamide), not “in the range of 3.0 to 6.0,” as required by amended claim 1.

In sum, Masayoshi does not teach a method for obtaining a diamine from a diamide wherein the NaOX/diamide ratio is in the range of 3.0 to 6.0, as required by claim 1, and Andrews does not cure Masayoshi of this deficiency. Thus, the combination of Masayoshi and Andrews does not render the method of amended claim 1 obvious. Nor does their combination render obvious claims 2-4 and 7-10, which depend directly or indirectly from amended claim 1.

Even if a prima facie case of obviousness had been established based on Masayoshi and Andrews (which Applicants do not concede), it can be successfully rebutted by the unexpected results discussed above. More specifically, Examples 1-3 in the Specification and the example of the attached declaration (“Exhibit A”), where NaOX/diamide ratios were within the range of 3.0 to 6.0, and significant improvements

⁴ This reaction is also shown in the Specification at page 9, line 22, but for chlorine, rather than bromine.

in diamine yield and purity were obtained, compared with reactions in which the NaOX/diamide ratios were below this range (e.g., 2.0 to 2.5).

As discussed above, amended claims 5 and 11 each cover a method for obtaining a polyamic acid in which the first step in each method is identical to that of amended claim 1. Thus, for the same reasons and facts set forth above for claim 1, amended claims 5 and 11 are also not rendered obvious by the combination of Masayoshi and Andrews. Nor are claims 6 and 12 which depend from claims 5 and 11, respectively.

III

To complete the record, Applicants would like to address the Examiner's assertion that "[Andrews] teaches that the NaOH to diamide (NaOH/diamide ratio) is 6.0." The Examiner errs in his calculation of the NaOH/diamine molar ratio by dividing 75 mmols by the molar amount of diamide of 12 mmols to arrive at the ratio of 6.0. The correct NaOH/diamide ratio is 2.25, not 6.0. More specifically, the correct ratio of 2.25 can be obtained by dividing the moles of NaOH upon completion of the reaction, i.e., 27 mmols, by the moles of diamide, i.e., 12.0 mmols. Admittedly, the NaOH/diamide ratio of 2.25, disclosed in Andrews, is within the range recited in amended claim 1. However, the patentability of claim 1 resides in part in "a NaOX/diamine ratio within the range of 3.0-6.0," not "a NaOH/diamine ratio within the range of 1.8-6.0."

CONCLUSION

It is believed that all of the pending claims have been addressed. However, the absence of a reply to a specific rejection, issue or comment does not signify agreement with or concession of that rejection, issue or comment.

In addition, because the arguments made above may not be exhaustive, there may be reasons for patentability of any or all pending claims (or other claims) that have not been expressed.

Finally, nothing in this paper should be construed as an intent to concede any issue with regard to any claim, except as specifically stated in this paper, and the

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amendment of any claim does not necessarily signify concession of unpatentability of the claim prior to its amendment.

No fees are believed due. Please apply any other charges or credits to Deposit Account No. 50-4189, referencing Attorney Docket No. 60004-109US1.

Respectfully submitted,

Date: 2-13-08


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